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A THERMAL EQUATION FOR FLAME QUENCHING

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SUMMARY

An approximate thermal equation was derived for quenching distance based on a previously proposed diffusional treatment. The quenching distance is expressed in terms of the thermal conductivity, the fuel mole fraction, the heat capacity, the rate of the rate-controlling chemical reactions, a constant that depends on the geometry of the quenching surface, and one empirical constant.

In order to use the equation with experimental data, the rate-controlling reaction must be specified. Two choices were made: (1) the reaction between active particles and fuel (corresponding to the diffusional equation), and (2) the reaction between oxygen and fuel.

The effect of pressure on quenching distance was shown to be inversely proportional to the pressure dependence of the primary flame reaction, with a small correction necessitated by the effect of pressure on flame temperature.

The general equation was used with the Semenov equation for burning velocity to show that the quenching distance was inversely proportional to burning velocity and pressure at any given initial temperature and equivalence ratio.

The two equations, one based on the active-particle - fuel reaction, the other based on the oxygen-fuel reaction, were tested, using published data for the quenching of propane-oxygen-nitrogen flames. The data selected included the effect on quenching distance of oxygen-nitrogen ratio, propane concentration, and initial mixture temperature and pressure. A correlation of these data was obtained, using each of the two equations, although both possessed shortcomings. The equation using the active-particle - fuel reaction did not correlate data for all rich mixtures; the equation using the oxygen-fuel reaction, while correlating data for both rich and lean mixtures, showed a larger deviation from the predicted linear relation than the other equation.

INTRODUCTION

The process of flame quenching is of interest, since it may be related to other combustion phenomena of engineering importance, such as flame stabilization, flammability limits, and the general behavior of flames near cold walls. Flame-quenching processes become especially important in turbojet combustion systems when operation at low pressure (i.e., high altitudes) is considered. Flame quenching is usually studied experimentally in terms of the quenching distance, which is defined as the minimum channel size that will allow a given flame to propagate. Most of the available quenching data have been obtained either by (1) observation of the minimum tube diameter or rectangular slot width that will allow a flame to flash back, or by (2) determination of the minimum distance between plane-parallel plates that will allow a flame to propagate from a spark of minimum ignition energy.

A rigorous theoretical treatment of the quenching process appears to be a most difficult task (refs. 1 and 2). Consequently, present theoretical treatments of quenching are necessarily approximate and seek primarily to correlate the quenching process with the variables that affect it, such as pressure, temperature, fuel type and concentration, inert-gas concentration, and quenching-surface geometry.

Approximate treatments of quenching have been based either on a thermal or a diffusional mechanism for the process (refs. 3 and 4). Such apparently different outlooks yield results consistent with experimental data because the equations for heat and mass transfer are formally identical, and because the thermodiffusivity and the molecular diffusivity are numerically nearly equal (ref. 5). Thus, each purely thermal approach to a combustion process has a diffusional analog.

A quenching-distance equation that successfully correlates lean quenching data (refs. 4, 6, and 7) is the diffusional equation given by Simon, Belles, and Spakowski (ref. 4). Unfortunately, the use of this equation in its present form is limited to stoichiometric or lean mixtures. The source of this difficulty appears to be the choice of the reaction kinetics.

Essentially, the objective of this paper is to extend, if possible, the useful range of the quenching concepts of reference 4 to include hydrocarbon-rich mixtures. Because of the similarities between heat and mass flow, it is to be expected that a thermal analog equation can be derived that will correlate data equally as well as the diffusional equation of reference 4. It was believed that such an alternate thermal equation might be more susceptible to changes in the reaction kinetics than the original diffusional model. It was hoped that the result of such changes would be an extension of the useful range of the equation to include rich mixtures.

This report contains the derivation of a thermal quenching equation in which the rate-controlling reaction is not specified. Two possible rate-controlling reactions are postulated, and the resulting two quenching equations are tested using published quenching data. It is shown that, if the oxygen-fuel reaction is assumed to be rate-controlling, quenching data are satisfactorily correlated for both rich and lean propane-oxygen-nitrogen flames. The pressure dependence of the quenching distance and the relation of quenching distance to burning velocity are also discussed.

THEORY

As an initial assumption, the authors of reference 4 assumed that in order for a flame to propagate the number of reaction events per cubic centimeter that occur in the gas ahead of the burning zone (hereinafter called the reaction zone) must be above some minimum value. The number of reaction events per cubic centimeter in the reaction zone is assumed to be the number of active particles per cubic centimeter c_i multiplied by the average number of effective collisions ν_i (the average active-particle chain length) made by each particle before its destruction at the wall. The minimum number of reaction events necessary for flame propagation is assumed to be a constant fraction A of the total number of molecules per cubic centimeter N_T . In terms of these definitions, the criterion for quenching is given as

$$\nu_i c_i \leq A N_T \quad (1)$$

for a single species of active particle and a surface chain-breaking efficiency of 1 (symbols are defined in the appendix).

The definition of the number of reaction events necessary for flame propagation used in reference 4 is not a unique one. Another possibility is to assume that the necessary number of reaction events in the reaction zone is a constant fraction F of the total number of events per cubic centimeter x that normally occur during the passage of a flame through a unit volume of gas. The quenching criterion then becomes

$$\nu_i c_i \leq F x \quad (2)$$

This quenching criterion leads to a more suitable thermal equation than would be obtained through the use of equation (1).

Equation (2) may be converted to its thermal counterpart by multiplying through by hRT/P to give as a final result (expressed in cal/mole):

$$H_r \leq FH_T \quad (3)$$

Here, the number of calories per mole produced in the reaction zone H_r must be equal to or less than some critical value, which is assumed to be a constant fraction F of the total heat produced per mole of unburned gas H_T .

For the combustion of hydrocarbons, complete combustion of the fuel is assumed to occur for all mixture compositions. Consequently, the total heat produced by the process may be written as the product of the mole fraction of fuel in the unburned gas X_F and the heat produced upon the disappearance of 1 mole of fuel ΔH (as defined herein, ΔH is constant and equal to the heat of combustion to CO_2 and H_2O only for stoichiometric or lean mixtures; for rich mixtures, ΔH decreases as the oxygen concentration decreases). Thus, for hydrocarbon flames, equation (3) may be written as

$$H_r \leq FX_F\Delta H \quad (4)$$

The expression given in reference 4 for the chain length ν_i is

$$\nu_i = \frac{d^2}{G_i D_i \tau_i} \quad (5)$$

The thermal equivalent of this equation is found by multiplying through by $hc_i RT/P$ to obtain as a final result

$$H_r = \frac{d^2 hc_i RT}{G_i D_i \tau_i P} \quad (6)$$

According to the definitions given in reference 4,

$$\frac{c_i}{\tau_i} = ka_r c_i = w \quad (7)$$

The heat produced per reaction event in the reaction zone is proportional to the heat produced upon the disappearance of one fuel molecule. For simplicity, the two are assumed to be equal so that the heat produced per reaction event may be written as

$$h = \frac{\Delta H}{N} \quad (8)$$

Since the molecular diffusivity and thermodiffusivity are assumed to be equivalent,

$$D_i = \frac{\alpha_r RT}{PC_{p,r}} \quad (9)$$

Any small error implicit in the assumption of equation (9) should not influence the over-all behavior of the resulting equation for quenching distance.

Combination of equations (6) to (9) yields the thermal analog of equation (5):

$$H_r = \frac{d^2 \Delta H C_{p,r} w}{G_i N \alpha_r} \quad (10)$$

Simultaneous solution of equations (4) and (8) gives for the thermal quenching equation

$$d^2 = \frac{FG_i N \alpha_r X_f}{C_{p,r} w} \quad (11)$$

(The inequality sign has been removed since the quenching distance is related to the tube configuration that just quenches a given flame.) The most important fact concerning this equation is that the form for w is no longer specified; any reaction may now be chosen as rate-controlling.

If equation (1) rather than equation (2) had been used as the quenching criterion, the final thermal quenching equation would have been formally identical with equation (11), except that the fuel mole fraction X_f would not appear. This difference is unimportant as regards the correlation of data if only a narrow range of fuel concentration is considered. For data that cover a wide range of fuel concentration, the thermal quenching equation as written in equation (11) is found to be the more satisfactory of the two possibilities.

In terms of the active-particle - fuel reaction (the reaction specified as rate-controlling in the diffusional equation of ref. 4), the reaction rate w is given by

$$w = k_t a_r \sum_i c_{i,r} \quad (12)$$

If a collision-theory-type temperature dependence is assigned to k_t , equation (12) becomes

$$w = B_t a_r \sum_i c_{i,r} T_r^{1/2} \exp\left(\frac{-E_t}{RT_r}\right) \quad (13)$$

Combination of equations (10) and (13) gives

$$d^2 = \frac{FG_i N \alpha_r X_f}{C_{p,r} B_t a_r \sum_i c_{i,r} T_r^{1/2} \exp\left(\frac{-E_t}{RT_r}\right)} = \frac{F}{B_t} \psi_t \quad (14)$$

The choice of rate-controlling reaction is not limited to the active-particle - fuel reaction. For example, it is possible to follow Semenov (ref. 8) and assume the reaction in the combustion zone to be bimolecular and first-order with respect to fuel and oxygen. For this assumption, the real reacting species are obviously not fuel and oxygen molecules, but such an assumption is satisfactory if the concentrations of the reacting species are proportional to the fuel and oxygen concentrations. In this case, then, the rate of the reaction is given by

$$w = k_s a_r b_r \quad (15)$$

If a collision-theory-type temperature dependence is assigned to k_s , equation (15) becomes

$$w = B_s a_r b_r T_r^{1/2} \exp\left(\frac{-E_s}{RT_r}\right) \quad (16)$$

The combination of equations (10) and (16) gives

$$d^2 = \frac{FG_i N \alpha_r X_f}{C_{p,r} B_s a_r b_r T_r^{1/2} \exp\left(\frac{-E_s}{RT_r}\right)} = \frac{F}{B_s} \psi_s \quad (17)$$

In order to test equations (14) and (17), data for the quenching of propane-oxygen-nitrogen flames were used. These data include the effect on quenching distance of oxygen-nitrogen ratio, fuel concentration and pressure (ref. 6), and initial temperature (ref. 9).

In order to use equation (14), the assumptions made in reference 4 concerning the active-particle - fuel reaction were followed: T_r was taken to be $0.7 T_F$, the partial pressure of fuel in the reaction zone

was assumed to be one-half the partial pressure of fuel in the unburned gas, the partial pressures of the active particles (H atoms, OH radicals, and O atoms) in the reaction zone were assumed to be 0.7 times their partial pressure in the adiabatic equilibrium flame and E_t was assumed to be 7 kilocalories per mole (ref. 10).

In order to use equation (17), the partial pressures of fuel and oxygen in the reaction zone were taken as equal to their values in the unburned gas and E_s was assumed to be 38 kilocalories per mole (ref. 11). In keeping with previous usage (refs. 4 and 6), T_r was chosen to be equal to $0.7 T_F$.

The thermal conductivities were calculated for the unburned gas mixture at $0.7 T_F$. The conductivities for oxygen, nitrogen, and propane were calculated at 1553°K , using the tables and data given in chapter 8 of reference 12. Conductivities at other temperatures were computed on the assumption of a $3/4$ -power temperature dependence. The thermal conductivities of the gas mixtures were calculated as the sum of the mole fraction times the thermal conductivity of each component, or,

$$\kappa = \sum_i X_i \kappa_i \quad (18)$$

It was felt that this simple linear mixing rule was adequate for systems largely composed of nitrogen and oxygen because of their similar conductivities.

The heat capacities were calculated for the unburned gas mixture at $0.7 T_F$ by the same linear mixing rule used for the thermal conductivities. Individual heat capacities for oxygen, nitrogen, and propane were calculated from data given in reference 13.

The equilibrium adiabatic flame temperatures and product compositions were calculated by the matrix method of reference 14 using the thermodynamic constants of reference 14 and the heat of formation of propane given in reference 13.

RESULTS AND DISCUSSION

Correlation of Quenching-Distance Data

Inasmuch as the square of the quenching distance is predicted to be directly proportional to ψ_t (eq. (14)) or to ψ_s (eq. (17)), values of ψ_t and ψ_s were calculated for the pressure, equilibrium adiabatic flame composition and temperature, unburned gas composition and temperature, and tube geometry (assumed to be plane-parallel plates), which

correspond to the quenching distances reported in references 6 and 9 for propane-oxygen-nitrogen flames. Plots of d^2 against ψ_t and ψ_s are shown in figures 1(a) and (b), respectively. The ranges of the variables included were: pressure, 0.1 to 1.0 atmosphere; equivalence ratio, 0.33 to 1.90; mole fraction oxygen in the oxidant mixture, 0.21 to 0.70; and unburned gas temperature, 300° to 558° K. In figure 1(a), it is seen that ψ_t (which is entirely analogous to the diffusional equation of ref. 4 except for the choice of quenching criterion) satisfactorily correlates the effect of these variables except for propane-rich mixtures where $\phi > 1.2$.

Inspection of figure 1(b) reveals that ψ_s (which includes the oxygen-fuel reaction as the rate-determining step, rather than the active-particle - fuel reaction) correlates not only the propane-lean, but also all the propane-rich quenching data. Thus, it appears that by proper choice of the rate-controlling reaction, the quenching concepts of reference 4 may be extended in such a way as to include rich mixtures for the propane-oxygen-nitrogen system.

A mean line drawn through the data presented in figure 1 reveals that the predicted linear relation between d^2 and ψ_t and ψ_s does not hold exactly. Rather, the data are best correlated if $d^2 \propto (\psi_t)^{0.94} \propto (\psi_s)^{0.84}$. This difficulty is related, at least in part, to the choice of reaction mechanism, since the "best correlation" exponent is changed by a change in reaction mechanism.

Pressure Dependence of Quenching Distance

Examination of the equation for quenching distance (eq. (11)) reveals that all terms on the right side are independent of pressure except the reaction rate w . However, hydrocarbon flame temperatures change slightly with pressure because of changes in the extent of dissociation. Consequently, a pressure change indirectly affects temperature- and composition-dependent terms in the equation. This effect is quite small, so that the pressure dependence of the quenching distance may be regarded as primarily dependent on the pressure dependence of the initial chemical processes in the combustion wave.

The calculated pressure dependence of the quenching distance (including the indirect effect of pressure on flame temperature and composition) is compared in table I with the experimental values of reference 6. The average deviation of the calculated values from the observed values was found to be 10 percent for ψ_t and 17 percent for ψ_s .

Reference 6 gives a value of 7 percent for the average deviation of values computed for the diffusional equation.

It is seen that ψ_s does not predict the pressure dependence as well as does ψ_t , except for the high-velocity flames at an oxygen fraction of 0.70. This is probably because the assumption of a simple second-order reaction between fuel and oxygen is a poor one. Concerning prediction of pressure dependence, a better choice of over-all order would be 1.7 rather than 2.0; this is in agreement with theoretical studies of the pressure dependence of burning velocity (ref. 12, p. 765), which indicate that the global reaction in most hydrocarbon flames ranges between first and second order. Such a choice would also tend to improve the linearity of the relation between d^2 (observed) and ψ_s (calculated).

Quenching Distance and Burning Velocity

The relation of quenching distance to burning velocity has been discussed in references 3, 4, and 15. In reference 4, an equation relating the two variables is derived on a purely diffusional basis. The thermal analog to this equation may be derived as follows:

According to reference 16, the Semenov equation for the burning velocity may be written as

$$U^2 = \frac{2\alpha_F \bar{W}}{a_o \rho_o \bar{C}_{p,F}} \left(\frac{n_1}{n_2} \right)^m \left(\frac{\lambda RT}{DC_p P} \right)_F^m \quad (19)$$

Multiplying equation (11) by equation (19) and writing for a_o and ρ_o ,

$$a_o = \frac{NX_F P}{RT_o} \quad (20a)$$

$$\rho_o = \frac{\bar{M}P}{RT_o} \quad (20b)$$

yields

$$d = \frac{RT_o}{UP} \sqrt{\frac{2\alpha_r \alpha_F F G_i \left(\frac{n_1}{n_2} \right)^m \left(\frac{\lambda RT}{DC_p P} \right)_F^m \bar{W}}{C_{p,r} \bar{C}_{p,F} \bar{M} w}} \quad (21)$$

If it is assumed that \bar{W} and w have the same pressure dependence, it follows from equation (21) that, at a given equivalence ratio,

$$U \propto \frac{1}{dP} \propto P^{-(n+1)} \quad (22)$$

From reference 6, it is seen that n decreases from about -0.9 to -1.05 as the percent oxygen in the oxidant mixture is increased from 21 to 70. Simultaneously, the burning velocity increases from about 40 centimeters per second to the order of 300 centimeters per second. Consequently, equation (22) predicts that the exponent describing the pressure dependence of U should increase from about -0.1 to 0.05 as U increases from 40 centimeters per second to about 300 centimeters per second. This result may be compared with the experimental findings of Lewis (ref. 17) (which indicate an increase of about -0.05 to 0.07 as burning velocity changes from 40 to 300 cm/sec) to confirm equation (22) qualitatively.

CONCLUDING REMARKS

The fact that reasonable agreement exists between experiment and quenching concepts based on either heat or mass transfer indicates only that these processes have the same formal laws and that the thermal and molecular diffusivities are either equal or remain directly proportional for the data considered in this study. No evidence for the preponderance of either heat or mass transfer in quenching can be deduced from these results. The principal advantage gained by formulation of the quenching concepts of reference 4 in a "thermal" form is that a change in reaction kinetics is much simpler from a conceptual point of view for the "thermal" form than for the "diffusional" form. This conceptual advantage might be useful in a discussion of bizarre fuel-oxidant systems for example, CS_2 - F_2 flames. In addition, the actual calculations may be simplified, since by the proper choice of reaction mechanism, it may become unnecessary to calculate the equilibrium flame composition.

SUMMARY OF RESULTS

The results of an investigation into the possibility of extending quenching concepts previously proposed to include quenching of hydrocarbon-rich flames may be summarized as follows:

1. An equation for the critical quenching configuration of a channel was derived for hydrocarbon flames:

$$d^2 = \frac{FG_1 N \kappa_r X_f}{C_{p,r} w}$$

where

- $C_{p,r}$ heat capacity in reaction zone, cal/(°K)(mole)
- d characteristic dimension of tube geometry; quenching distance, cm
- F constant that relates total number of reaction events that occur during the passage of a flame to number that must occur in the reaction zone for flame propagation
- G_1 dimensionless geometrical factor, dependent only on channel geometry
- N Avogadro's number
- w rate of reaction in reaction zone, molecules/(cc)(sec)
- X_f mole fraction of fuel in unburned gas
- κ_r mean thermal conductivity in reaction zone, cal/(cm)(sec)(°K)

2. Two possible flame-initiating reactions were considered in the detailed formulation of w : (1), the reaction between active particles and fuel molecules, and (2), the reaction between oxygen and fuel molecules.

3. The two equations resulting from the two reaction mechanism choices were tested using published data that included the effect of oxygen-nitrogen ratio, fuel concentration, pressure, and unburned gas temperature on the quenching distance for propane-oxygen-nitrogen flames. These variables were correlated reasonably well for both rich and lean flames by the equation involving the oxygen-fuel reaction. The equation involving the active-particle - fuel reaction was satisfactory primarily for lean flames.

4. It was concluded that, by the proper choice of reaction kinetics, the quenching concepts previously proposed may be cast in a form useful for the prediction of both propane-rich and -lean quenching data.

Lewis Flight Propulsion Laboratory
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APPENDIX - SYMBOLS

The following symbols are used in this report:

A	fraction of molecules present that must react for flame to continue to propagate
a	fuel concentration, molecules/cc
B	Arrhenius constant
b	oxygen concentration, molecules/cc
c_i	concentration of i^{th} active particle, molecules/cc
C_p	heat capacity, cal/(°K)(mole)
$\bar{C}_{p,F}$	average heat capacity, T_0 to T_F , cal/(°K)(mole)
D	diffusion coefficient, cm^2/sec
d	characteristic dimension of tube geometry; quenching distance, cm
E	activation energy, cal/mole
F	constant that relates the total number of reaction events that occur during the passage of a flame to the number which must occur in the reaction zone for flame propagation
G_i	dimensionless factor, value of which is dependent only on tube geometry
ΔH	heat released upon consumption of 1 mole of fuel by combustion process, cal/mole
H_r	heat produced by chemical reaction in primary reaction zone, cal/mole
H_T	total heat produced by combustion of 1 mole of unburned gas, cal/mole
h	heat produced per reaction event, cal
k	rate constant, (cc)(molecules)/sec
\bar{M}	average molecular weight of unburned gas, g/mole
m	molecularity of flame reaction

N	Avogadro's number
N_T	total number of molecules present per cc
n	exponent describing pressure dependence of quenching distance
n_1/n_2	moles of reactant per moles of product from stoichiometric equation
P	pressure, atm
R	gas constant
T	temperature, °K
U	flame speed, cm/sec
\bar{W}	average reaction rate in flame front as defined by Semenov, molecules/(cc)(sec)
w	reaction rate in primary reaction zone, molecules/(cc)(sec)
X_f	mole fraction of fuel
x	number of reaction events per cc that occur during passage of flame through a unit volume
α	mole fraction of oxidant in oxidant-inert mixture
κ	thermal conductivity, cal/(cm)(sec)(°K)
ν	average active-particle chain length
ρ	density, g/cc
τ	time between effective collisions, sec
ϕ	equivalence ratio

$$\psi_s = \frac{G_i N \kappa_r X_f}{C_{p,r} a_r b_r T_r^{1/2} \exp\left(\frac{-E_s}{RT_r}\right)}$$

$$\psi_t = \frac{G_i N \kappa_r X_f}{C_{p,r} a_r \sum_i c_{i,r} T_r^{1/2} \exp\left(\frac{-E_t}{RT_r}\right)}$$

Subscripts:

F	flame
i	active-particle species
o	unburned gas
r	primary reaction zone
s	involves reaction of oxygen and fuel molecules
t	involves reaction of active particles and fuel molecules

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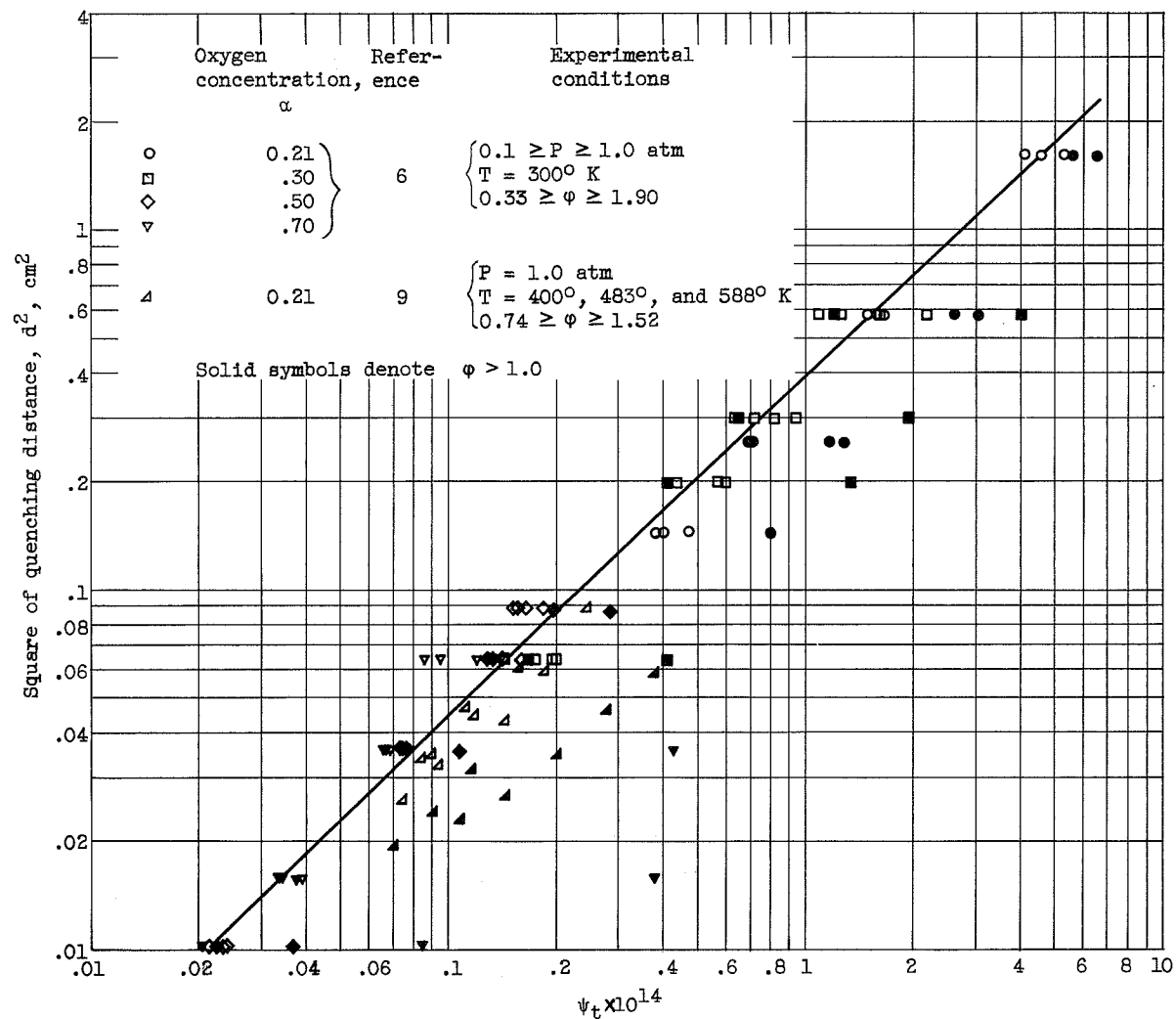
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TABLE I. - COMPARISON OF OBSERVED PRESSURE DEPENDENCE
OF QUENCHING DISTANCE WITH PREDICTED VALUES

Oxygen fraction, α	Equivalence ratio, ϕ	Exponent n describing pressure dependence, $d \propto p^{-n}$			
		Observed ^a	Predicted		
			Diffu- sional ^a	Thermal	
				ψ_t	ψ_s
0.17	0.943	0.90	0.88	0.83	1.03
	1.000	.89	.85	.76	1.01
	1.340	.84	.75	.75	1.00
	1.530	.71	.75	.75	1.00
0.21	0.738	0.85	0.86	0.80	1.00
	.864	.84	.89	.84	1.03
	1.000	.89	.93	.89	1.03
	1.240	.95	.78	.79	1.02
	1.490	.98	.76	.77	1.00
0.30	0.566	0.76	0.87	0.87	1.01
	.662	.93	.89	.88	1.04
	.773	1.06	.92	.90	1.07
	1.000	.98	.94	.90	1.09
	1.380	.93	.86	.84	1.06
	1.903	.74	.76	.76	1.01
0.50	0.476	1.01	0.91	0.89	1.06
	.544	1.01	.93	.91	1.07
	.680	.96	.96	.92	1.09
	1.000	.93	.97	.92	1.11
	1.358	.91	.95	.90	1.09
	1.940	.88	.82	.81	1.05
0.70	0.345	1.12	0.91	0.89	1.06
	.395	1.12	.93	.91	1.07
	.494	1.02	.96	.92	1.09
	1.000	1.01	.98	.93	1.09
	1.234	1.07	.98	.92	1.11
	1.829	.60	.89	.86	1.08

^aRef. 6.



(a) Quenching distance against ψ_t .

Figure 1. - Summary correlations of quenching data by thermal quenching equations (data from refs. 6 and 9).

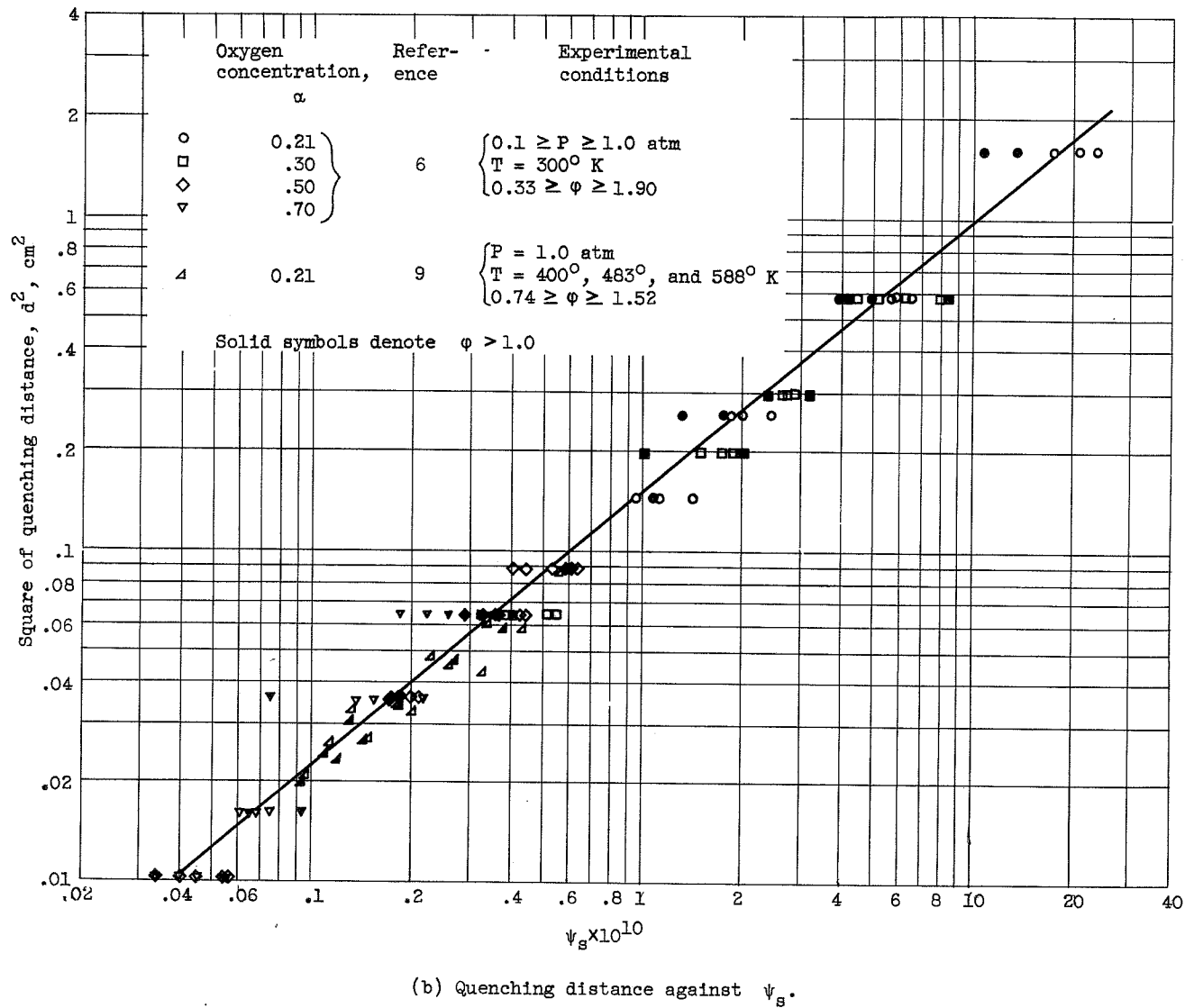


Figure 1. - Concluded. Summary correlations of quenching data by thermal quenching equations (data from refs. 6 and 9).

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A thermal quenching equation is derived; the equation is essentially an extension of a previously proposed diffusional concept. By proper choice of the rate-controlling chemical reaction, the equation becomes suitable for use with rich as well as lean fuel-air mixtures. The equation was tested, using published quenching-distance data for propane-oxygen-nitrogen flames, which include the effect of oxygen-nitrogen ratio, equivalence ratio, pressure, and initial temperature.

Copies obtainable from NACA, Washington

1. Fuels - Properties, Physical and Chemical (3.4.2)
 2. Combustion Research - General (3.5.1)
 3. Combustion - Reaction Mechanisms (3.5.1.5)
- I. Potter, A. E., Jr.
 - II. Berlad, Abraham L.
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